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An Unconditionally Gradient Stable Adaptive Mesh Refinement for the Cahn-Hilliard Equation

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We consider a numerical method, the so-called an unconditionally gradient stable adaptive mesh refinement scheme, for solving the Cahn-Hilliard equation representing a model of phase separation in a binary mixture. The continuous problem has a decreasing total energy. We show the same property for the corresponding discrete problem by using eigenvalues of the Hessian matrix of the energy functional. An unconditionally gradient stable time discretization is used to remove the high-order time-step constraints. An adaptive mesh refinement is used to highly resolve narrow interfacial layers.

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I. INTRODUCTION

The Cahn-Hilliard (CH) equation [1] was originally introduced as a phenomenological model of phase separation in a binary alloy. It is a leading model in theoretical materials science and it has been applied to a wide range of problems. Examples of its applications are phase separation [2], multiphase fluid flow [3–6], flow visualization [7], image processing [8], a morphological instability caused by elastic non-equilibrium [9] and the formation of the quantum dots [10] (see Refs. [11,12,27–29] for the physics of quantum dots). Therefore, an efficient and accurate numerical solution of the equation is needed to understand its dynamics. We consider an unconditionally gradient stable adaptive mesh refinement algorithm for the CH equation:

$$\frac{\partial c(\mathbf{x},t)}{\partial t} = M\Delta\mu(c(\mathbf{x},t)), \quad \mathbf{x}\in\Omega,\ 0<t\le T, \tag{1}$$

$$\mu(c(\mathbf{x},t)) = F'(c(\mathbf{x},t)) - \epsilon^2\Delta c(\mathbf{x},t), \tag{2}$$

where $\Omega \subset \mathbf{R}^d$ $(d = 1, 2, 3)$ is a domain. The quantity $c(\mathbf{x}, t)$ is defined to be the difference between the respective concentrations of the two mixtures' components. The coefficient M is a constant mobility and we take $M \equiv 1$ for convenience. The function $F(c)$ is the bulk free energy of a homogeneous solution. The small constant ϵ is the gradient energy coefficient, which is related to the interfacial energy. The boundary conditions on $\partial\Omega$ are

$$\frac{\partial c}{\partial \mathbf{n}} = \frac{\partial \mu}{\partial \mathbf{n}} = 0 \text{ on } \partial\Omega, \tag{3}$$

where $\mathbf{n}$ is the unit vector normal to $\partial\Omega$ and $\frac{\partial}{\partial \mathbf{n}}$ denotes the normal derivative on $\partial\Omega$. The physical meaning of the first condition is that the total free energy of the mixture decreases in time. The meaning of the second one is that none of the mixture can pass through the walls of the container and is equivalent to $\frac{\partial \Delta c}{\partial \mathbf{n}} = 0$. The CH equation arises from the Ginzburg-Landau free energy

$$\mathcal{E}(c) := \int_\Omega \left[F(c) + \frac{\epsilon^2}{2}|\nabla c|^2\right] d\mathbf{x}. \tag{4}$$

We consider the bulk free energy as a double well potential of the form $F(c) = \frac{1}{4}(c^2-1)^2$ as in Ref. [13]. We differentiate the energy $\mathcal{E}$ and the total mass $\int_\Omega c d\mathbf{x}$ in the sense of Gâteaux to get

$$\begin{aligned}\frac{d}{dt}\mathcal{E}(c) &= \int_\Omega (F'(c)c_t + \epsilon^2\nabla c\cdot\nabla c_t)d\mathbf{x}\\ &= \int_\Omega \mu c_t d\mathbf{x} = \int_\Omega \mu\Delta\mu d\mathbf{x}\\ &= \int_{\partial\Omega}\mu\frac{\partial\mu}{\partial\mathbf{n}}ds - \int_\Omega \nabla\mu\cdot\nabla\mu d\mathbf{x} = -\int_\Omega |\nabla\mu|^2 d\mathbf{x} \le 0\end{aligned} \tag{5}$$

and

$$\frac{d}{dt}\int_\Omega c d\mathbf{x} = \int_\Omega c_t d\mathbf{x} = \int_\Omega \Delta\mu d\mathbf{x} = \int_{\partial\Omega}\frac{\partial\mu}{\partial\mathbf{n}}ds = 0, \tag{6}$$

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where we have used the boundary conditions in Eq. (3). Therefore, the total energy is non-increasing in time; that is, the total energy is a Lyapunov functional for solutions of the Cahn-Hilliard equation. Furthermore, the total mass is conserved,

$$\int_{\Omega} c(\mathbf{x},t)d\mathbf{x} = \text{constant}. \tag{7}$$

Numerical simulations of the Cahn-Hilliard equation are difficult on a normal computer in a reasonable time because both the biharmonic operator and the nonlinear operator impose severe time-step restrictions on using explicit methods ($\Delta t \sim h^4$), so implicit methods must be used. Additionally, across the spatial interfaces, the solution undergoes an $O(1)$ change over an $O(\epsilon)$ interval. If these interfaces are to be accurately resolved, a fine discretization of space is required. Therefore, an adaptive mesh refinement of the space is necessary. Ideally, one would like to use a stable integration algorithm which would allow accuracy requirements rather than stability limitations to determine the integration step size. Then, very small time step sizes, which cause huge computational costs and make the calculation inefficient, are required in order to use a reasonable time step size, Eyre introduced a concept "unconditionally gradient stable scheme," in Refs. [14,15].

We use an unconditionally gradient stable scheme with an adaptive mesh refinement to solve the resulting discrete equations accurately and efficiently. In Eyre's papers [14,15], he provided the idea and the theory of the scheme a little vaguely. We here provide mathematical reasoning for the scheme. We emphasize that while the methods will allow us to take arbitrarily large time steps, the accuracy of the numerical solution depends on choosing a small enough time step to resolve the dynamics [15].

This paper is organized as follows: In Section II, we briefly review a derivation of the Cahn-Hilliard equation. This derivation is based on constrained gradient dynamics for a physically motivated functional. In Section III, we describe the discrete scheme and its properties, such as mass conservation and total energy, decrease. We present the numerical results in Section IV. Section V contains a discussion.

II. THE CAHN-HILLIARD EQUATION

In this section, we review the Cahn-Hilliard equation. We follow the analysis in Refs. [16,17]. When a binary alloy is rapidly quenched from a melt to a low temperature, the homogeneous mixture usually becomes unstable. A pattern of domain formation, called spinodal decomposition, takes places as the two metals within the alloy separate out. From the initial phase of spinodal decomposition, the mixture quickly becomes inhomogeneous, forming a fine-grained structure that exhibits a characteristic length scale. After that, a slow coarsening process can be observed while the above-mentioned characteristic length scale grows.

In a series of papers [1,18,19], Cahn and Hilliard analyzed these processes mentioned above and they proposed the Cahn-Hilliard equation. The derivative of the bulk free energy $F'(c) = c(c^2-1)$ has zeros $c_0 = 0, c_1 = -1$ and $c_2 = 1$. $F''(c)$ has zeroes $c_1^* = -\frac{1}{\sqrt{3}}$ and $c_2^* = \frac{1}{\sqrt{3}}$. $F''(c) > 0$ for $c < c_1^*$ or $c > c_2^*$, $F''(c) < 0$ for $c_1^* < c < c_2^*$. The intervals (c_1, c_1^*), (c_1^*, c_2^*) and (c_2^*, c_2) are called metastable interval 1, the spinodal interval, and metastable interval 2, respectively [17].

Constant functions $c = c_m$ are equilibria for Eq. (1). These functions model the homogeneous mixture. Such functions are unstable if c_m is in the spinodal interval, that is, if $F''(c_m) < 0$. Such uniform mixture $c = c_m$ is then very unstable and the growth of instabilities results in phase separation, which is called spinodal decomposition.

Now, we review a derivation of the Cahn-Hilliard equation as a gradient flow. It is natural to seek a law of evolution in the form

$$\frac{\partial c}{\partial t} = -\text{grad}_0 \mathcal{E}(c). \tag{8}$$

The symbol "grad_0" here denotes the gradient on the manifold, in the sense of the Gâteaux derivative in a Hilbert space, defined by Eq. (7). Let the domain $\mathcal{D}$ of the functional $\mathcal{E}$ be the set of smooth enough functions c defined in Ω and satisfying $\frac{\partial c}{\partial \mathbf{n}} = 0$. Let M_0 be the linear manifold in $\mathcal{D}$ of functions c satisfying $\int_\Omega c d\mathbf{x} = 0$. Given any $c \in \mathcal{D}$, define $M_c = c + M_0$. Because of Eq. (7) and considering $c - (c)_\Omega$, we may consider functions with zero average. Here, $(c)_\Omega = \frac{1}{|\Omega|}\int_\Omega c$ means the average of c over Ω.

For any Hilbert space H, we denote H^* as its dual space. Denote by $\dot{H}^{-1}$ the zero-average subspace of the dual $(H^1)^*$ of the usual Hilbert space $H^1 = W^{1,2}(\Omega)$. Let $\dot{H}^1$ be the zero-average subspace of H^1, with norm $\|\nabla u\|_{L^2}$ and inner product $(u,v)_{\dot{H}^1} \equiv (\nabla u, \nabla v)_{L^2}$ for $u, v \in \dot{H}^1$. The inner product of $u, v \in \dot{H}^{-1}$ is defined by

$$(u,v)_{\dot{H}^{-1}} \equiv (\nabla \phi_u, \nabla \phi_v)_{L^2},$$

where $\phi_u, \phi_v \in \dot{H}^1$ are the associates of u, v. For example [20–22], ϕ_u satisfy

$$\Delta \phi_u = u \quad \text{in } \Omega, \quad \frac{\partial \phi_u}{\partial \mathbf{n}} = 0 \quad \text{on } \partial\Omega, \quad \int_\Omega \phi_u d\mathbf{x} = 0.$$

Then, grad_0 is defined in the following way: Let $\dot{C}_0^\infty(\Omega)$ be the set of smooth functions with compact support and with zero average. Then, $\dot{C}_0^\infty$ is dense in $\dot{H}^{-1}$. Let c be sufficiently smooth and satisfy $\frac{\partial c}{\partial \mathbf{n}} = \frac{\partial \Delta c}{\partial \mathbf{n}} = 0$ on $\partial\Omega$. Then, we have that for all $v \in \dot{C}_0^\infty$,

$$(\text{grad}_0 \mathcal{E}(c), v)_{\dot{H}^{-1}} = \frac{d}{d\theta}\mathcal{E}(c + \theta v)\big|_{\theta=0}$$

$$= \lim_{\theta\to 0}\frac{1}{\theta}\big(\mathcal{E}(c+\theta v)-\mathcal{E}(c)\big)$$
$$= \int_\Omega \big(F'(c)-\epsilon^2\Delta c\big)v d\mathbf{x}. \quad (9)$$

Put $\Delta\phi_v$ in place of v in Eq. (9). An integration by parts then yields

$$\int_\Omega [F'(c)-\epsilon^2\Delta c]\Delta\phi_v d\mathbf{x}$$
$$= -\int_\Omega \nabla[F'(c)-\epsilon^2\Delta c]\cdot\nabla\phi_v d\mathbf{x} \quad (10)$$
$$= (-\nabla[F'(c)-\epsilon^2\Delta c],\nabla\phi_v)_{L^2} \quad (11)$$
$$= (-\nabla\cdot\nabla[F'(c)-\epsilon^2\Delta c],\nabla\cdot\nabla\phi_v)_{\dot{H}^{-1}} \quad (12)$$
$$= (-\Delta[F'(c)-\epsilon^2\Delta c],v)_{\dot{H}^{-1}} \quad (13)$$

because $\nabla\phi_v$ has zero normal component on $\partial\Omega$. On the space $\dot{H}^{-1}$, the Ginzburg-Landau free energy $\mathcal{E}$ is defined by Eq. (4). We identify

$$\mathrm{grad}_0\mathcal{E}(c) \equiv -\Delta[F'(c)-\epsilon^2\Delta c] \quad (14)$$

and specify its domain as those functions in $\mathcal{D}$ that satisfy Eq. (3) [17].

III. NUMERICAL ANALYSIS

In this section, we present a fully discrete scheme for the solution of the CH equation. In addition, we prove discrete versions of mass conservation and energy dissipation, which immediately imply the stability of the numerical scheme. We consider an unconditionally gradient stable scheme for time discretization introduced in Eyre [14,15] and show that the discrete total free energy is nonincreasing for any time step size Δt. For simplicity of exposition, we shall discretize the CH equation, Eqs. (1) and (2), in one-dimensional space, *i.e.*, $\Omega=(a,b)$. Two and three-dimensional discretizations are defined analogously.

Let N be a positive even integer, $h=(b-a)/N$ be the uniform mesh size and $\Omega_h=\{x_i=(i-0.5)h,\ 1\le i\le N\}$ be the set of cell-centers. Let c_i^n and μ_i^n be approximations of $c(x_i,n\Delta t)$ and $\mu(x_i,n\Delta t)$, respectively, where Δt is the time step. We first implement the zero Neumann boundary condition, Eq. (3), by requiring that for each n,

$$\nabla_h c^n_{\frac12}=\nabla_h c^n_{N+\frac12}=\nabla_h\mu^n_{\frac12}=\nabla_h\mu^n_{N+\frac12}=0, \quad (15)$$

where the discrete differentiation operator is $\nabla_h c^n_{i+\frac12}=(c^n_{i+1}-c^n_i)/h$. We then define a discrete Laplacian by $\Delta_h d_i=(\nabla_h d_{i+\frac12}-\nabla_h d_{i-\frac12})/h$ and discrete l_2 inner product by

$$\langle\mathbf{d},\mathbf{e}\rangle_h = h\sum_{i=1}^N d_i e_i \ \text{ and}$$
$$(\nabla_h\mathbf{d},\nabla_h\mathbf{e})_h = h\sum_{i=0}^N \nabla_h d_{i+\frac12}\nabla_h e_{i+\frac12},$$

where $\mathbf{d}=(d_1,d_2,\cdots,d_N)$, $\mathbf{e}=(e_1,\cdots,e_N)$ and $\nabla_h\mathbf{d}=(\nabla_h d_{\frac12},\nabla_h d_{\frac32},\cdots,\nabla_h d_{N+\frac12})$. We also define the discrete norm as $||\mathbf{c}||_h^2=\langle\mathbf{c},\mathbf{c}\rangle_h$. Then, using the boundary condition Eq. (15), we have

$$\langle\Delta_h\mathbf{d},\mathbf{e}\rangle_h=\langle\mathbf{d},\Delta_h\mathbf{e}\rangle_h=-(\nabla_h\mathbf{d},\nabla_h\mathbf{e})_h. \quad (16)$$

For dissipative dynamics such as the CH equation, a discrete time stepping algorithm is defined to be *unconditionally gradient stable* if the discrete total free energy is nonincreasing for *any* size of a time step Δt.

Eyre's theorem [15] shows that an unconditionally gradient stable algorithm results for the CH equation if we can split the free energy appropriately into contractive and expansive parts,

$$\mathcal{E}(c)=\int_a^b\left[F(c)+\frac{\epsilon^2}{2}c_x^2\right]dx \quad (17)$$
$$=\int_a^b\left[\frac{c^4+1}{4}+\frac{\epsilon^2}{2}c_x^2\right]dx-\int_a^b\frac{c^2}{2}dx$$
$$=\mathcal{E}_c(c)-\mathcal{E}_e(c),$$

and then treat the contractive part $\mathcal{E}_c(c)$ implicitly and the expansive part $-\mathcal{E}_e(c)$ explicitly. We use the nonlinearly stabilized splitting scheme [15] that involves a semi-implicit time and centered difference space discretizations of Eqs. (1) and (2):

$$\frac{c_i^{n+1}-c_i^n}{\Delta t}=\Delta_h\mu_i^{n+\frac12}\ \text{for } i=1,\cdots,N, \quad (18)$$
$$\mu_i^{n+\frac12}=\nu_i^{n+1}-c_i^n, \quad (19)$$
$$\nu_i^{n+1}=(c_i^{n+1})^3-\epsilon^2\Delta_h c_i^{n+1}. \quad (20)$$

The main purpose of this section is to show that the scheme in Eqs. (18)-(20) inherits the characteristic properties, has a decrease in the total energy and maintains the conservation of mass corresponding to Eqs. (6) and (6). Let $\boldsymbol{c}^n=(c_1^n,c_2^n,\cdots,c_N^n)$ and $\boldsymbol{\mu}^n=(\mu_1^n,\mu_2^n,\cdots,\mu_N^n)$ and $\mathbf{1}=(1,1,\cdots,1)$; then, the second assertion follows by using a discrete summation by parts and the boundary condition Eq. (15):

$$\langle\mathbf{c}^{n+1},\mathbf{1}\rangle_h=\langle\mathbf{c}^n,\mathbf{1}\rangle_h+\Delta t\langle\Delta_h\boldsymbol{\mu}^{n+\frac12},\mathbf{1}\rangle_h$$
$$=\langle\mathbf{c}^n,\mathbf{1}\rangle_h-\Delta t(\nabla_h\boldsymbol{\mu}^{n+\frac12},\nabla_h\mathbf{1})_h=\langle\mathbf{c}^n,\mathbf{1}\rangle_h. \quad (21)$$

To show the decrease in the discrete total energy, first we define a discrete Lyapunov functional,

$$\mathcal{E}^h(\mathbf{c}^n)=\frac{h}{4}\sum_{i=1}^N((c_i^n)^2-1)^2+\frac{\epsilon^2h}{2}\sum_{i=0}^N|\nabla_h c^n_{i+\frac12}|^2. \quad (22)$$

for each n. It is useful to break $\mathcal{E}^h(\mathbf{c}^n)$ into three parts:

$$\mathcal{E}^{(1)}(\mathbf{c}^n)=-\frac{h}{2}\sum_{i=1}^N(c_i^n)^2,$$
$$\mathcal{E}^{(2)}(\mathbf{c}^n)=\frac{\epsilon^2h}{2}\sum_{i=0}^N\left|\nabla_h c^n_{i+\frac12}\right|^2, \text{ and}$$

$$\mathcal{E}^{(3)}(\mathbf{c}^n) = \frac{h}{4}\sum_{i=1}^{N}\left((c_i^n)^4+1\right).$$

We define a decomposition of $\mathcal{E}^h(\mathbf{c}^n)$ as $\mathcal{E}_c^h(\mathbf{c}^n) = \mathcal{E}^{(2)}(\mathbf{c}^n)+\mathcal{E}^{(3)}(\mathbf{c}^n)$ and $\mathcal{E}_e^h(\mathbf{c}^n) = -\mathcal{E}^{(1)}(\mathbf{c}^n)$. We define grad_h as the minus of the Laplacian of the variational derivative with respect c_i^n; *i.e.*,

$$\begin{aligned}\mathrm{grad}_h\mathcal{E}^h(\mathbf{c}^n)_i &= -\Delta_h\frac{\delta\mathcal{E}^h(\mathbf{c}^n)}{\delta c_i^n}\\ &= -\Delta_h((c_i^n)^3 - c_i^n - \epsilon^2\Delta_h c_i^n)\\ &= -\Delta_h(c_i^n)^3 + \Delta_h c_i^n + \epsilon^2\Delta_h^2 c_i^n, \end{aligned}\tag{23}$$

where the discrete biharmonic operator $\Delta_h^2 c_i$ is defined as $\Delta_h(\Delta_h c_i)$. We can rewrite the numerical scheme in Eqs. (18)-(20) in terms of a gradient of the discrete total energy; *i.e.*,

$$\frac{c_i^{n+1}-c_i^n}{\Delta t} = -\mathrm{grad}_h\mathcal{E}_c^h(\mathbf{c}^{n+1})_i + \mathrm{grad}_h\mathcal{E}_e^h(\mathbf{c}^n)_i,\quad \text{for } i=1,\cdots,N.\tag{24}$$

The Hessian of $\mathcal{E}^{(1)}(\mathbf{c})$, denoted by $\mathbf{H}^{(1)}$, is the Jacobian of $\mathrm{grad}_h\mathcal{E}^{(1)}(\mathbf{c})$ and is thus given by

$$\begin{aligned}\mathbf{H}^{(1)} &= J\mathrm{grad}_h\mathcal{E}^{(1)}(\mathbf{c})\\ &= \frac{1}{h^2}\begin{pmatrix} -1 & 1 & & & 0\\ 1 & -2 & 1 & & \\ & \ddots & \ddots & \ddots & \\ & & 1 & -2 & 1\\ 0 & & & 1 & -1\end{pmatrix},\end{aligned}$$

the Hessian matrix of $\mathcal{E}^{(2)}(\mathbf{c})$ is given as

$$\begin{aligned}\mathbf{H}^{(2)} &= J\mathrm{grad}_h\mathcal{E}^{(2)}(\mathbf{c})\\ &= \frac{\epsilon^2}{h^4}\begin{pmatrix} 2 & -3 & 1 & & & & 0\\ -3 & 6 & -4 & 1 & & & \\ 1 & -4 & 6 & -4 & 1 & & \\ & \ddots & \ddots & \ddots & \ddots & \ddots & \\ & & 1 & -4 & 6 & -4 & 1\\ & & & 1 & -4 & 6 & -3\\ 0 & & & & 1 & -3 & 2\end{pmatrix},\end{aligned}$$

and the Hessian matrix of $\mathcal{E}^{(3)}(\mathbf{c})$ is given as

$$\begin{aligned}\mathbf{H}^{(3)} &= J\mathrm{grad}_h\mathcal{E}^{(3)}(\mathbf{c})\\ &= \frac{3}{h^2}\begin{pmatrix} c_1^2 & -c_2^2 & & & 0\\ -c_1^2 & 2c_2^2 & -c_3^2 & & \\ & \ddots & \ddots & \ddots & \\ & & -c_{N-2}^2 & 2c_{N-1}^2 & -c_N^2\\ 0 & & & -c_{N-1}^2 & c_N^2\end{pmatrix}\\ &= -3\mathbf{H}^{(1)}\begin{pmatrix} c_1^2 & 0 & & & 0\\ 0 & c_2^2 & 0 & & \\ & \ddots & \ddots & \ddots & \\ & & 0 & c_{N-1}^2 & 0\\ 0 & & & 0 & c_N^2\end{pmatrix},\end{aligned}$$

where we have used the boundary condition in Eq. (15). The eigenvalues of $\mathbf{H}^{(1)}$, $\mathbf{H}^{(2)}$ and $\mathbf{H}^{(3)}$ are

$$\lambda_k^{(1)} = -\frac{4}{h^2}\sin^2\frac{(k-1)\pi}{2N},\quad k=1,2,\cdots,N,\tag{25}$$

$$\lambda_k^{(2)} = \epsilon^2(\lambda_k^{(1)})^2 = \frac{16\epsilon^2}{h^4}\sin^4\frac{(k-1)\pi}{2N},\tag{26}$$

$$0 \le \lambda_k^{(3)} \le \frac{12}{h^2}\sin^2\frac{(N-1)\pi}{2N}\max_{1\le i\le N}c_i^2.\tag{27}$$

Note that $\lambda_k^{(1)}$ is non-positive and that $\lambda_k^{(2)}$ and $\lambda_k^{(3)}$ are non-negative. Let $\mathbf{v}_i$ for $i=1,\cdots,N$ be common orthonormal eigenvectors of $\mathbf{H}^{(1)}$ and $\mathbf{H}^{(2)}$ corresponding to the eigenvalues, $\lambda_i^{(1)}$ and $\lambda_i^{(2)}$, respectively. Let $\lambda_1^e,\lambda_2^e,\ldots,\lambda_N^e$ be eigenvalues of $J(\mathrm{grad}_h\mathcal{E}_e^h) = -\mathbf{H}^{(1)}$; *i.e.*,

$$\lambda_k^e = -\lambda_k^{(1)} = \frac{4}{h^2}\sin^2\frac{(k-1)\pi}{2N},\quad k=1,2,\cdots,N.\tag{28}$$

We can expand $\mathbf{c}^{n+1}-\mathbf{c}^n$ in the basis of eigenvalues $\mathbf{v}_i$ as follows.

$$\mathbf{c}^{n+1}-\mathbf{c}^n = \sum_{k=1}^{N}\alpha_k\mathbf{v}_k.\tag{29}$$

The decrease of the discrete energy functional is established in the following theorem: If $\mathbf{c}^{n+1}$ is the solution of Eqs. (18) - (20) with given $\mathbf{c}^n$, then

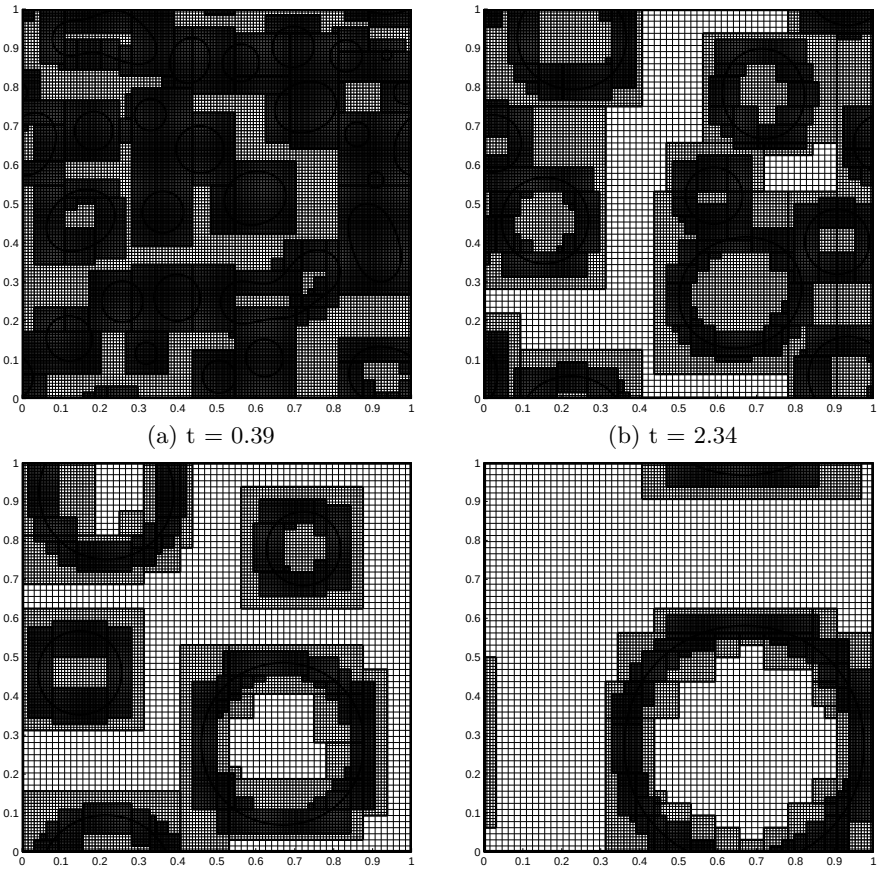
$$\mathcal{E}^h(\mathbf{c}^{n+1}) \le \mathcal{E}^h(\mathbf{c}^n).\tag{30}$$

Next, we prove Eq. (30). With an exact Taylor expansion of $\mathcal{E}^h(\mathbf{c}^n)$ about $\mathbf{c}^{n+1}$ up to second order, we have

$$\begin{aligned}\mathcal{E}^h(\mathbf{c}^n) &= \mathcal{E}^h(\mathbf{c}^{n+1}) + \langle\mathrm{grad}_h\mathcal{E}^h(\mathbf{c}^{n+1}),\mathbf{c}^n-\mathbf{c}^{n+1}\rangle_h\\ &\quad+\left\langle\frac{J(\mathrm{grad}_h\mathcal{E}^h)(\boldsymbol{\xi})}{2}(\mathbf{c}^n-\mathbf{c}^{n+1}),\mathbf{c}^n-\mathbf{c}^{n+1}\right\rangle_h,\end{aligned}$$

where $\boldsymbol{\xi} = \theta\mathbf{c}^n+(1-\theta)\mathbf{c}^{n+1}$, $0\le\theta\le1$. Rearranging the terms and using $\mathcal{E}^h = \mathcal{E}^{(1)}+\mathcal{E}^{(2)}+\mathcal{E}^{(3)}$, Eq. (29), Eq. (24) and the mean value theorem, we have

$$\begin{aligned}&\mathcal{E}^h(\mathbf{c}^{n+1})-\mathcal{E}^h(\mathbf{c}^n)\\ &= \left\langle\mathrm{grad}_h\mathcal{E}^h(\mathbf{c}^{n+1}) - \frac{J(\mathrm{grad}_h\mathcal{E}^h)(\boldsymbol{\xi})}{2}\right.\\ &\left.\times(\mathbf{c}^{n+1}-\mathbf{c}^n),\mathbf{c}^{n+1}-\mathbf{c}^n\right\rangle_h\\ &\le \left\langle\mathrm{grad}_h\mathcal{E}^h(\mathbf{c}^{n+1}) - \frac{1}{2}(\mathbf{H}^{(1)}+\mathbf{H}^{(2)})\right.\\ &\left.\times(\mathbf{c}^{n+1}-\mathbf{c}^n),\mathbf{c}^{n+1}-\mathbf{c}^n\right\rangle_h\\ &= \left\langle\mathrm{grad}_h\mathcal{E}^h(\mathbf{c}^{n+1}),\mathbf{c}^{n+1}-\mathbf{c}^n\right\rangle_h\\ &\quad-\sum_{j,k=1}^{N}\left\langle\frac{1}{2}(\mathbf{H}^{(1)}+\mathbf{H}^{(2)})\alpha_j\mathbf{v}_j,\ \alpha_k\mathbf{v}_k\right\rangle_h\\ &= \left\langle\mathrm{grad}_h\mathcal{E}_c^h(\mathbf{c}^{n+1}) - \mathrm{grad}_h\mathcal{E}_e^h(\mathbf{c}^{n+1})\right.\\ &\left.-\frac{1}{\Delta t}(\mathbf{c}^{n+1}-\mathbf{c}^n) - \mathrm{grad}_h\mathcal{E}_c^h(\mathbf{c}^{n+1})\right.\end{aligned}$$



(a) t = 0.39 (b) t = 2.34

(c) t = 7.42 (d) t = 39.06

Fig. 1. Evolution of the concentration $c(x, y, t)$ with an average concentration $c_m = -0.4$. The times are shown below each figure. The effective fine grid resolution for 2 levels of adaptivity is 256×256.

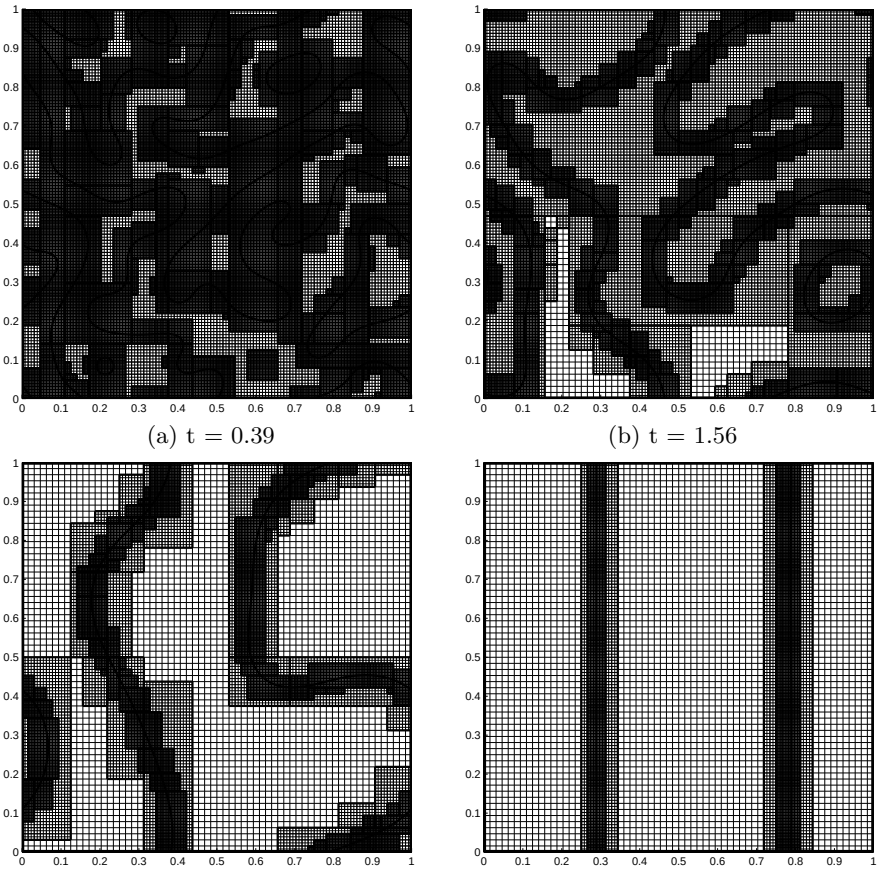
$$
\begin{aligned}
&+\mathrm{grad}_h\mathcal{E}_e^h(\mathbf{c}^n), \mathbf{c}^{n+1} - \mathbf{c}^n\Big\rangle_h \\
&- \sum_{j,k=1}^{N} \Big\langle \frac{1}{2}(\lambda_j^{(1)} + \lambda_j^{(2)})\alpha_j \mathbf{v}_j,\ \alpha_k \mathbf{v}_k \Big\rangle_h \\
&= -\Big\langle \mathrm{grad}_h\mathcal{E}_e^h(\mathbf{c}^{n+1}) - \mathrm{grad}_h\mathcal{E}_e^h(\mathbf{c}^n),\ \mathbf{c}^{n+1} - \mathbf{c}^n \Big\rangle_h \\
&- \frac{1}{\Delta t}\|\mathbf{c}^{n+1} - \mathbf{c}^n\|_h^2 \\
&- \sum_{j,k=1}^{N} \Big\langle \frac{1}{2}(\lambda_j^{(1)} + \lambda_j^{(2)})\alpha_j \mathbf{v}_j,\ \alpha_k \mathbf{v}_k \Big\rangle_h \\
&= -\sum_{j,k=1}^{N} \Big\langle \Big[J(\mathrm{grad}_h\mathcal{E}_e^h) + \frac{1}{2}(\lambda_j^{(1)} + \lambda_j^{(2)})I\Big] \\
&\times \alpha_j \mathbf{v}_j,\ \alpha_k \mathbf{v}_k \Big\rangle_h - \frac{1}{\Delta t}\|\mathbf{c}^{n+1} - \mathbf{c}^n\|_h^2 \\
&= -\sum_{k=1}^{N} \frac{1}{2}\big(\lambda_k^e + \lambda_k^{(2)}\big)\alpha_k^2 - \frac{1}{\Delta t}\|\mathbf{c}^{n+1} - \mathbf{c}^n\|_h^2 \le 0,
\end{aligned}
$$

where we have used the fact that λ_k^e and $\lambda_k^{(2)}$ are both non-negative. Therefore, we have proven the decrease of the discrete total energy. This completes the proof. The theorem holds for any time step Δt; hence, the method is unconditionally gradient stable.

There are many authors (for example, see Refs. [13,23, 24]) who cited Ref. [14]. Note that the proof in Ref. [14] is vague even though the theorem itself is true. There are a lot of counter examples for which the claim, "all of the eigenvalues of $J(\mathrm{grad}_h\mathcal{E}_e^h)$ dominate the largest eigenvalues of $-J(\mathrm{grad}_h\mathcal{E}^h)$" does not hold. For example, let us consider $N = 10$, $h = 1/N$, $\epsilon = 2h$ and $\mathbf{c} = \mathbf{0}$; then, the eigenvalues of $J(\mathrm{grad}_h\mathcal{E}_e^h)$ are $\frac{4}{h^2}\sin^2\frac{(k-1)\pi}{20}$ for $k = 1, 2, \cdots, 10$. In particular, when $k = 1$, an eigenvalue is zero. On the other hand, the eigenvalues of $-J(\mathrm{grad}_h\mathcal{E}^h)$ are $\frac{4}{h^2}\sin^2\frac{(k-1)\pi}{20} - \frac{16\epsilon^2}{h^4}\sin^4\frac{(k-1)\pi}{20}$ for $k = 1, 2, \cdots, 10$. When $k = 2$, one of eigenvalues of $-J(\mathrm{grad}_h\mathcal{E}^h)$ is $\frac{4}{h^2}\sin^2\frac{\pi}{20} - \frac{16\epsilon^2}{h^4}\sin^4\frac{\pi}{20} \approx 5.95595$, which is greater than zero.

IV. NUMERICAL EXAMPLES

We validate our method via spinodal decomposition, mass conservation and an energy decrease. We implement the unconditionally gradient stable scheme in Eqs.



(a) t = 0.39 (b) t = 1.56

(c) t = 5.47 (d) t = 39.06

Fig. 2. Evolution of the concentration $c(x, y, t)$ with an average concentration $c_m = 0$. The times are shown below each figure. The effective fine grid resolution for 2 levels of adaptivity is 256×256.

(18)-(20) with the recently developed adaptive mesh refinement methodology. For a detailed description of the numerical method used in solving these equations, please refer to Refs. [25,26].

1. Spinodal Decomposition

The initial state is taken to be $c(x, y, 0) = c_m + 0.05\text{rand(x,y)}$ on the computational domain $\Omega = [0, 1] \times [0, 1]$. The average concentration is c_m and rand(x,y) is a random number between -1 and 1. We use the simulation parameters $\epsilon = 0.005$ and $\Delta t = 1/256$. It should be emphasized that while the methods will allow us to take arbitrarily large time steps, the accuracy of the numerical solution depends on choosing a small enough time step to resolve the fast-time-scale dynamics. We also use a base 64×64 mesh with two levels of refinement. Therefore, the effective fine mesh size is 256×256. For the sharp interface limit on a unit square domain and double periodic boundary conditions, it can be shown that if $|c_m| > 1-2/\pi$, then the minimum energy is achieved by a single circular domain. If $|c_m| < 1 - 2/\pi$, the minimum energy configuration consists of two straight interfaces parallel to either the x or the y axis. Figure 1 shows an evolution of the concentration $c(x, y, t)$ with an average concentration $c_m = -0.4$ at times $t = 0.39,\ 2.34,\ 7.42$ and 39.06. Figure 2 shows the evolution of the concentration $c(x, y, t)$ with an average concentration $c_m = 0$ at times $t = 0.39,\ 1.56,\ 5.47$ and 39.06. We note that in the limit of the sharp interface, figure 1(d) and figure 2(d) are the global minimizers of the total free energy for the average concentrations $c_m = -0.4$ and $c_m = 0$, respectively.

2. The Decrease of the Total Energy and Conservation of Mass

In Figure 3, the time evolution of the non-dimensional discrete total energy $\mathcal{E}^h(\mathbf{c}^n)/\mathcal{E}^h(\mathbf{c}^0)$ (solid line: $c_m = -0.4$ and dashed-dot line: $c_m = 0$) and the average concentrations (diamond: $c_m = -0.4$ and circle: $c_m = 0$) of the previous numerical experiments are shown. Also, the inscribed small figures are the concentration fields at indicated times. The energies are non-increasing and the

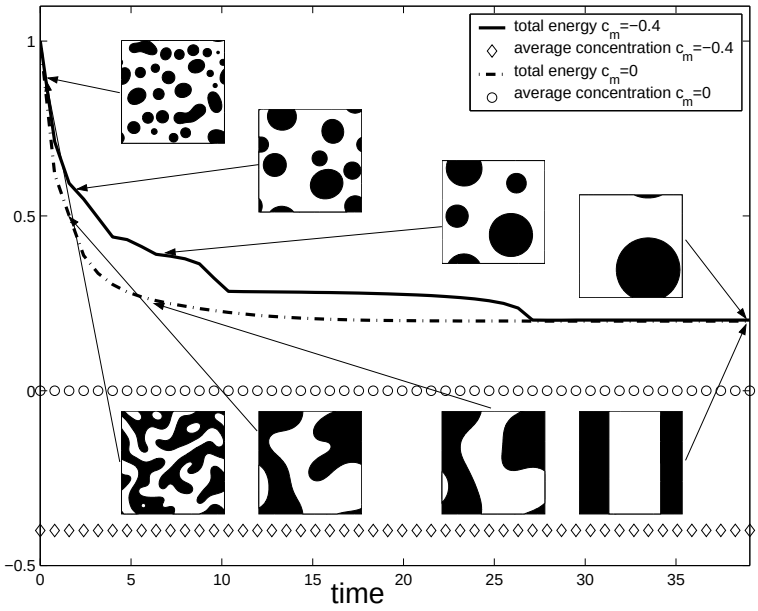


Fig. 3. Non-dimensional discrete total energy $\mathcal{E}^h(\mathbf{c}^n)/\mathcal{E}^h(\mathbf{c}^0)$ (solid line: $c_m = -0.4$ and dashed-dot line: $c_m = 0$) and the average concentrations (diamond: $c_m = -0.4$ and circle: $c_m = 0$) of the numerical solutions.

average concentrations are conserved. These numerical results agree well with the total energy dissipation property in Eq. (30) and the conservation property in Eq. (21).

V. CONCLUSIONS

In this paper, we briefly reviewed the Cahn-Hilliard equation and by using eigenvalues of the Hessian matrix of the energy functional, we showed explicitly that the scheme in Eqs. (18)-(20) is an unconditionally gradient stable scheme. Thus, we clarified the details of the proof. In particular, there are a few equations in Ref. [15] that lack factors of vector norms. More substantively, we found that the proof of Eyre's theorem as originally presented was slightly ambiguous and abstractive to follow. An adaptive mesh methodology was used to focus the computational effort to regions near the interfacial transition zone.

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